IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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In re Application of Group Art Unit: 1795

Makoto HIGAMI, et al. Examiner: Helen O. Chu

Application No. 10/776,313

Filed: 02/12/2004

For: ELECTRODE PASTE COMPOSITION

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

DECLARATION UNDER 37 CFR 1.132

I, Junji KAWAI, declare and state that:

1. In March, 1999, I was graduated from the graduate course of Kyoto Institute of Technology, majoring in Polymer and received a degree of Master of Engineering.

Since April, 1999, I have been an employee of JSR CORPORATION, and I have been engaged in the research and development work concerning energy field.

2. I am a co-inventor of the invention described in the specification of the above-identified application.

3. The following Experiments were carried out in order to demonstrate the superiority of the presently claimed invention.

[Comparative Example 5]

<Preparation of paste H>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

0.22 g of platinum-supporting carbon particles (Pt content: 46 wt%);

2.55 g of distilled water;

6.14 g of a water-alcohol solution containing 20.6 wt% Nafion (trade name, available from Dupont) (water:alcohol weight ratio = 20:60);

9.14 g of n-propyl alcohol;

5.76 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.46 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.036 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The above water-alcohol solution of Nafion contained ethanol and n-propyl alcohol as the alcohols.

The contents were stirred with a wave rotor for 70 minutes to give a paste H having a viscosity of 25 cp (25°C).

[Evaluation]

Electrodes and a fuel cell were produced by the procedure illustrated in Example 1 except that the paste H was used in place of the paste A. The properties were evaluated as described in Example 1. The results are shown in Tables 1 and 2.

[Comparative Example 6]

<Preparation of paste I>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

1.68 g of platinum-supporting carbon particles (Pt content: 46 wt%);

2.55 g of distilled water;

1.06 g of a water-alcohol solution containing 20.6 wt% Nafion (trade name, available from Dupont) (water:alcohol weight ratio = 20:60);

14.22 g of n-propyl alcohol;

4.3 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.46 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.036 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The above water-alcohol solution of Nafion contained ethanol and n-propyl alcohol as the alcohols.

The contents were stirred with a wave rotor for 70 minutes to give a paste I having a viscosity of 30 cp (25°C).

[Evaluation]

The paste I was applied on carbon paper in a platinum amount of 0.5 mg/cm^2 with use of a doctor blade. The resultant coating was dried at 95°C for 10 minutes to form an electrode layer, but the electrode layer was brittle and fell off carbon paper. Therefore, measurements of pore distribution and pore volume and production of fuel cell and evaluation of its properties could not be carried out, only evaluation of storage stability of electrode paste composition was carried out.

[Comparative Example 7]

<Preparation of paste J>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

1.68 g of platinum-supporting carbon particles (Pt content: 46 wt%);

2.55 g of distilled water;

6.14 g of a water-alcohol solution containing 20.6 wt% Nafion (trade name, available from Dupont) (water:alcohol weight ratio = 20:60);

2.0 g of n-propyl alcohol;

11.4 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.46 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.036 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The above water-alcohol solution of Nafion contained ethanol and n-propyl alcohol as the alcohols.

The contents were stirred with a wave rotor for 70 minutes to give a paste J having a viscosity of 100 cp (25°C).

[Evaluation]

The paste J was applied on carbon paper in a platinum amount of 0.5 mg/cm^2 with use of a doctor blade. The resultant coating was dried at 95°C for 10 minutes to form an electrode layer, but the electrode layer had no sufficient strength and it was difficult to maintain the shape on the carbon paper. Therefore, measurements of pore distribution and pore volume and production of fuel cell and evaluation of its properties could not be carried out, only evaluation of storage stability of electrode paste composition was carried out.

[Comparative Example 8]

<Preparation of paste K>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

6.00 g of platinum-supporting carbon particles (Pt content: 46 wt%);

2.55 g of distilled water;

6.14 g of a water-alcohol solution containing 20.6 wt% Nafion (trade name, available from Dupont) (water:alcohol weight ratio = 20:60);

4.85 g of n-propyl alcohol;

4.3 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.46 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.036 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The above water-alcohol solution of Nafion contained ethanol and n-propyl alcohol as the alcohols.

The contents were stirred with a wave rotor for 70 minutes to give a paste K having a viscosity of 400 cp (25°C).

[Evaluation]

Electrodes and a fuel cell were produced by the procedure illustrated in Example 1 except that the paste K was used in place of the paste A. The properties were evaluated as described in Example 1. The results are shown in Tables 1 and 2.

[Comparative Example 9]

<Preparation of paste L>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

1.25 g of platinum-supporting carbon particles (Pt content: 46 wt%);

1.89 g of distilled water;

16.34 g of a water-alcohol solution containing 50.0 wt% Nafion (trade name, available from Dupont) (water:alcohol weight ratio = 20:60);

2.97 g of n-propyl alcohol;

1.49 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.34 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.027 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The above water-alcohol solution of Nafion contained ethanol and n-propyl alcohol as the alcohols.

The contents were stirred with a wave rotor for 70 minutes to give a paste L having a viscosity of 900 cp (25°C).

[Evaluation]

Electrodes and a fuel cell were produced by the procedure illustrated in Example 1 except that the paste L was used in place

of the paste A. The properties were evaluated as described in Example 1. The results are shown in Tables 1 and 2.

[Comparative Example 10]

<Preparation of paste M>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

1.23 g of platinum-supporting carbon particles (Pt content: 46 wt%);

0.73 g of distilled water;

4.48 g of a water-alcohol solution containing 20.6 wt% Nafion (trade name, available from Dupont) (water:alcohol weight ratio = 20:60);

5.11 g of n-propyl alcohol;

12.40 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.34 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.026 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The above water-alcohol solution of Nafion contained ethanol and n-propyl alcohol as the alcohols.

The contents were stirred with a wave rotor for 70 minutes to give a paste M having a viscosity of 200 cp (25°C).

[Evaluation]

The paste M was applied on carbon paper in a platinum amount of 0.5 mg/cm^2 with use of a doctor blade. The resultant coating was dried at 95°C for 10 minutes to form an electrode layer, but the electrode layer was brittle and fell off carbon paper. Therefore, measurements of pore distribution and pore volume and production of fuel cell and evaluation of its properties could not be carried out, only evaluation of storage stability of electrode paste composition was carried out.

[Comparative Example 11]

<Preparation of paste N>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

0.90 g of platinum-supporting carbon particles (Pt content: 46 wt%);

0.54 g of distilled water;

3.29 g of a water-alcohol solution containing 20.6 wt% Nafion (trade name, available from Dupont) (water:alcohol weight ratio = 20:60);

18.77 g of n-propyl alcohol;

0.54 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.25 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.019 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The above water-alcohol solution of Nafion contained ethanol and n-propyl alcohol as the alcohols.

The contents were stirred with a wave rotor for 70 minutes to give a paste N having a viscosity of 30 cp (25°C).

[Evaluation]

The paste N was applied on carbon paper in a platinum amount of 0.5 mg/cm^2 with use of a doctor blade. The resultant coating was dried at 95°C for 10 minutes to form an electrode layer, but the electrode layer was brittle and fell off carbon paper. Therefore, measurements of pore distribution and pore volume and production of fuel cell and evaluation of its properties could not be carried out, only evaluation of storage stability of electrode paste composition was carried out.

[Comparative Example 12]

<Preparation of paste O>

A 50 ml glass bottle was charged with:

25 g of zirconia balls having 10 mm diameter (trade name: YTZ balls, available from Nikkato Corporation);

1.68 g of platinum-supporting carbon particles (Pt content: 46 wt%);

2.55 g of distilled water;

6.14 g of N,N-dimethylacetamide solution containing 20.6 wt% Nafion (trade name, available from Dupont);

9.14 g of N,N-dimethylacetamide and 4.3 g of n-butyl alcohol as the organic solvent of 100-200°C boiling point;

0.46 g of a vapor grown carbon fiber (trade name: VGCF, available from Showa Denko K.K.); and

0.036 g of a dispersant (trade name: DA234, available from Kusumoto Chemicals, Ltd.).

The contents were stirred with a wave rotor for 70 minutes to give a paste O having a viscosity of 150 cp (25°C).

[Evaluation]

Electrodes and a fuel cell were produced by the procedure illustrated in Example 1 except that the paste O was used in place of the paste A. The properties were evaluated as described in Example 1. The results are shown in Tables 1 and 2.

Table 1

	Paste	Additive solvent	Boiling point (°C)	Pore volume (ml/g)			Storage stability
				0.01-0.1 μm	0.1-1.0 μm	Total	
Comp. Ex. 5	H	n-butanol 24%	118	0.02	0.06	0.08	AA
Comp. Ex. 6	I	n-butanol 18%	118	*			AA
Comp. Ex. 7	J	n-butanol 47%	118	*			BB
Comp. Ex. 8	K	n-butanol 18%	118	0.20	0.50	0.70	AA
Comp. Ex. 9	L	n-butanol 6%	118	0.05	0.08	0.13	AA
Comp. Ex. 10	M	n-butanol 51%	118	*			BB
Comp. Ex. 11	N	n-butanol 2%	118	*			AA
Comp. Ex. 12	O	N,N-dimethyl acetamide 58%	166	0.10	0.20	0.30	AA
		n-butanol 18%	118				

*: no measurement

Table 2

Temperature and humidity		Low temperature and high humidity (50°C and 90% or more RH)		High temperature and high humidity (80°C and 90% or more RH)	
Current density (A/cm^2)		0.5	1.0	0.5	1.0
Voltage between the terminals (V)	Comp. Ex. 5	-	-	-	-
	Comp. Ex. 6	*			
	Comp. Ex. 7	*			
	Comp. Ex. 8	0.40	-	0.48	-
	Comp. Ex. 9	0.50	-	0.54	-
	Comp. Ex. 10	*			
	Comp. Ex. 11	*			
	Comp. Ex. 12	0.40	0.20	0.48	0.23

-: Unmeasurable due to the current density

*: no measurement

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Junji Kawai

Junji KAWAI

This 9th day of July, 2008